

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE PATENT APPLICATION OF :

Masayuki Ikeno et al : EXAMINER: ROBERTSON, JEFFREY

SERIAL No.: 10/020,219 :

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FOR: Organopolysiloxane composition for molding

\* \* \*  
CERTIFICATION OF TRANSLATION

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SIR:

I, Dr. Akinobu Abe, a technical engineer working with Iwamiya & Associates, Patent Attorneys having an office at NIPPON BUNJO-JUTAKU-KAIKAN BLDG, 3-6, KANDA OGAWA-MACHI, CHIYODA-KU, TOKYO, JAPAN, certify and declare:

(1) that I am fully conversant both with the Japanese and English languages;

(2) that I have carefully compared the priority document of Japanese Patent Application No. 2000-384154 in the Japanese language with an English translation thereof, a copy of said English translation being attached hereto;

(3) that the translation is, to the best my knowledge and belief, an accurate translation from the original into the English language; and

(4) that all statements made of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements are made with the knowledge that willful false statements and the like are punishable by fine or imprisonment, or both under 18 USC 1001, and that such false statements may jeopardize the validity of the application or any patent issuing thereon.

DATE: August 20, 2003 BY:

Akinobu Abe  
Akinobu Abe, Ph.D.

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**JAPAN PATENT OFFICE**

This is to certify that the annexed is a true copy of  
the following application as filed with this Office.

|                      |                              |
|----------------------|------------------------------|
| Date of Application: | December 18, 2000            |
| Application Number:  | 2000-384154                  |
| Applicant(s):        | Shin-Etsu Chemical Co., Ltd. |

August 24, 2001  
Kozo OIKAWA

(seal)  
Commissioner,  
Patent Office

Certification No.: 2001-3075966

[Document Name] Specification  
[Title of the Invention] Organopolysiloxane composition  
for molding

[Scope of Claim for Patent]

[Claim 1]

An organopolysiloxane composition for molding purposes comprising:

- (A) 100 parts by weight of an organopolysiloxane with at least two alkenyl groups bonded to silicon atoms within a single molecule, having a viscosity at 25°C of 0.05 to 100 Pa·s,
- (B) a straight chain organopolysiloxane with a hydrogen atom bonded to a silicon atom at both terminals and with no aliphatic unsaturated bonds within a molecule, having a viscosity at 25°C of 0.001 to 1.0 Pa·s,
- (C) an organohydrogenpolysiloxane with at least three hydrogen atoms bonded to silicon atoms within a single molecule and comprising a  $\text{RHSiO}$  unit and a  $\text{R}_2\text{XSiO}_{1/2}$  unit (wherein R is an unsubstituted or substituted monovalent hydrocarbon group with no alkenyl groups, and X represents a hydrogen atom or R) within a molecule as essential components, having a viscosity at 25°C of 0.001 to 1.0 Pa·s,
- (D) an effective quantity of a hydrosilylation reaction catalyst,
- (E) no more than 50 parts by weight of a finely powdered silica with a specific surface area of at least 50 m<sup>2</sup>/g, and
- (F) 0 to 20 parts by weight of a non-functional organopolysiloxane having a viscosity at 25°C of 0.01 to 500 Pa·s,

wherein a total number of hydrogen atoms bonded to

silicon atoms within said constituent (B) and said constituent (C) is in a range of 1 to 5 atoms per alkenyl group within said constituent (A), and a number of hydrogen atoms bonded to silicon atoms within said constituent (B) accounts for 20 to 70 mol% of a combined number of hydrogen atoms bonded to silicon atoms within said constituent (B) and said constituent (C).

[Detailed Description of the Invention]

[0001]

[Technical Field to Which the Invention Belongs]

The present invention relates to an organopolysiloxane composition for molding purposes effectively used as a molding reverse matrix, which upon curing displays superior mold releasability relative to urethane resins, epoxy resins, dicyclopentadiene resins, polyester resins, and the like.

[0002]

[Prior Art]

The production of a replica such as a urethane resin, an epoxy resin, a dicyclopentadiene resin or a polyester resin as a molding reverse matrix is a well-known technique. In recent years, resin molded products produced in this manner have been supplied for use in automobile components and household electrical components, and the characteristics of such resin molded products are being given serious consideration. As a result, the improvements in the characteristics of the casting resins have been quite dramatic, although unfortunately these improvements have resulted in a deterioration in the durability of the silicone rubber matrix, and the number of replica products that can be produced from a single matrix has decreased. Consequently, improvements in durability of the mold

releasability of the silicone rubber matrix with respect to these resins have been keenly sought.

[0003]

An improvement in mold release durability upon addition of an alkali metal hydroxide to a silicone composition is disclosed in Laid-open publication (kokai) No. 4-216864, whereas in Laid-open publication No. 5-279571, an improvement in mold releasability is disclosed for compositions incorporating a compound selected from the group consisting of an organotin compound, an organotitanium compound and an imidazole derivative. A technique for improving polyester mold durability using a radical scavenger is disclosed in Laid-open publication No. 11-158385. However, even with these modifications, the mold durability is still not entirely satisfactory. A composition using a polyfunctional cross-linking agent and a difunctional cross-linking agent is also disclosed in Laid-open publication No. 11-158385. However, the proportion of the difunctional cross-linking agent within the combined total of the cross-linking agents cannot be ascertained from the content of the above publication, and there is no indication that a combination of polyfunctional and difunctional chain extending agents produces superior mold release durability.

[0004]

[Problems the Invention Aims to Solve]

An object of the present invention is to provide an organopolysiloxane composition for molding purposes which displays superior mold releasability relative to urethane resins, epoxy resins, dicyclopentadiene resins, polyester resins, and the like.

[0005]

[Means for Solution of Problems]

The present invention discovered that by combining a polyfunctional cross-linking agent and a difunctional cross-linking agent as a chain lengthening agent within an organopolysiloxane composition, and moreover by ensuring that SiH groups within the difunctional cross-linking agent accounted for 20 to 70 mol% of SiH groups within the combined cross-linking agent, a composition could be produced which conformed to the above object, and as a result the present invention was completed.

[0006]

In other words, the present invention provides an organopolysiloxane composition for molding purposes comprising

- (A) 100 parts by weight of an organopolysiloxane with at least two alkenyl groups bonded to silicon atoms in a single molecule, having a viscosity at 25°C of 0.05 to 100 Pa·s,
- (B) a straight chain organopolysiloxane with a hydrogen atom bonded to a silicon atom at both terminals and with no aliphatic unsaturated bonds within the molecule, having a viscosity at 25°C of 0.001 to 1.0 Pa·s,
- (C) an organohydrogenpolysiloxane with at least three hydrogen atoms bonded to silicon atoms within a single molecule and substantially comprising a  $\text{RHSiO}$  unit and a  $\text{R}_2\text{XSiO}_{1/2}$  unit (wherein R is an unsubstituted or a substituted monovalent hydrocarbon group with no alkenyl groups, and X represents a hydrogen atom or R) within the molecule as essential components, having a viscosity at 25°C of 0.001 to 1.0 Pa·s,
- (D) an effective quantity of a hydrosilylation reaction catalyst.

(E) no more than 50 parts by weight of a finely powdered silica with a specific surface area of at least 50 m<sup>2</sup>/g, and

(F) 0 to 20 parts by weight of a non-functional organopolysiloxane having a viscosity at 25°C of 0.01 to 500 Pa·s, wherein

the total number of hydrogen atoms bonded to silicon atoms within the aforementioned constituent (B) and constituent (C) is in a range of 1 to 5 atoms per alkenyl group within the aforementioned constituent (A), and the number of hydrogen atoms bonded to silicon atoms within the constituent (B) accounts for 20 to 70 mol% of the combined number of hydrogen atoms bonded to silicon atoms within the constituent (B) and the constituent (C).

[0007]

[Embodiments of the Invention]

[Constituent (A)]

In the present invention, constituent (A) is an organopolysiloxane with at least two alkenyl groups bonded to silicon atoms in a single molecule. Specific examples of the alkenyl groups within the constituent (A) include vinyl groups, allyl groups, propenyl groups, isopropenyl groups, butenyl groups, isobutenyl groups, pentenyl groups, hexenyl groups and heptenyl groups, although vinyl groups are preferred. There are no particular restrictions on the bonding position of the alkenyl groups within the constituent (A), and molecular chain terminals and/or molecular side chains are suitable. Furthermore, examples of organic groups other than the alkenyl groups which may be bonded to the silicon atoms of the constituent (A) typically include unsubstituted or substituted monovalent hydrocarbon groups of 1 to 10 carbon atoms, and preferably

of 1 to 8 carbon atoms, with specific examples including alkyl groups such as methyl groups, ethyl groups, propyl groups, butyl groups, pentyl groups and hexyl groups; cycloalkyl groups such as cyclopentyl groups and cyclohexyl groups; aryl groups such as phenyl groups, tolyl groups, xylyl groups, and naphthyl groups; aralkyl groups such as benzyl groups and phenethyl groups; and halogen substituted alkyl groups such as 3,3,3-trifluoropropyl groups and 3-chloropropyl groups, although in terms of ease of synthesis, methyl groups are preferable.

[0008]

The viscosity at 25°C of the constituent (A) can be chosen from within a range from 0.05 to 100 Pa·s, with values from 0.1 to 30 Pa·s being preferable. The siloxane skeleton of the organopolysiloxane of the constituent (A) may be either a straight chain or a branched chain, or a mixture of the two, although a substantially straight chain diorganopolysiloxane in which the backbone chain comprises repeating diorganosiloxane units and both terminals of the molecular chain are blocked with a triorganosiloxy group is preferable.

[0009]

[Constituent (B)]

The constituent (B) used in the present invention is a straight chain organopolysiloxane having two SiH groups within the molecule, with a hydrogen atom bonded to a silicon atom at both terminals of the molecular chain (namely, SiH groups), and with no aliphatic unsaturated bonds within the molecule. The viscosity at 25°C of the constituent (B) is within a range from 0.001 to 1.0 Pa·s, with values from 0.01 to 0.1 Pa·s being preferable. This straight chain organopolysiloxane functions so as to

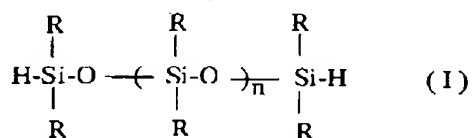


increase the molecular chain length of the aforementioned constituent (A) during the curing process, and has a significant effect on the molding durability.

An example of this organopolysiloxane can be represented by a general formula (I) shown below.

[0010]

[Chemical 1]



wherein, R is an unsubstituted or substituted monovalent hydrocarbon group with no alkenyl groups, and n is a number such that a viscosity at 25°C for the organopolysiloxane falls within the range described above.

[0011]

In the general formula (I), R represents an unsubstituted or substituted monovalent hydrocarbon group which incorporates no alkenyl groups, and is typically of 1 to 10 carbon atoms, and preferably of 1 to about 8 carbon atoms, with examples including alkyl groups such as methyl groups, ethyl groups, propyl groups, butyl groups, pentyl groups and hexyl groups; cycloalkyl groups such as cyclopentyl groups and cyclohexyl groups; aryl groups such as phenyl groups, tolyl groups, xylyl groups, and naphthyl groups; aralkyl groups such as benzyl groups and phenethyl groups; and halogen substituted alkyl groups such as 3,3,3-trifluoropropyl groups and 3-chloropropyl groups. n is a number such that a viscosity at 25°C for this organopolysiloxane falls within the aforementioned range

from 0.001 to 1.0 Pa·s, and preferably within a range from 0.01 to 0.1 Pa·s.

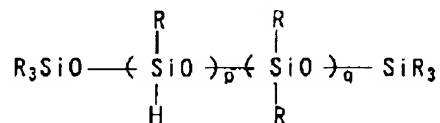
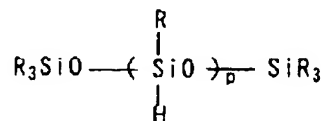
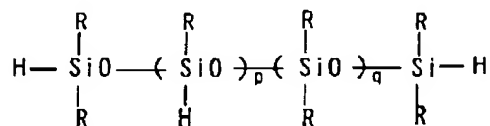
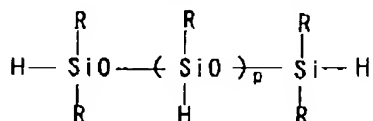
[0012]

[Constituent (C)]

In the present invention, constituent (C) is an organohydrogenpolysiloxane with at least three hydrogen atoms bonded to silicon atoms (SiH groups) within a single molecule. The hydrogen atoms bonded to silicon atoms may be positioned on terminal siloxane units and on siloxane units positioned within the polymer chain, or may be positioned only within the siloxane chain. This organohydrogenpolysiloxane is a straight chain siloxane polymer, incorporating units of general formulas  $\text{RHSiO}$  groups and  $\text{R}_2\text{XSiO}_{1/2}$  groups within the molecule as essential units, and may optionally incorporate units of  $\text{R}_2\text{SiO}$  groups. Examples are presented below.

[0013]

[Chemical 2]



wherein, R represents the same meaning as described above, p and q each represent an integer of 1 or greater, and p or p+q are values which satisfy the aforementioned viscosity.

[0014]

In these formulas, R represents the same as an unsubstituted or substituted monovalent hydrocarbon group as described for the general formula (I), which incorporates no alkenyl groups, and is typically of 1 to 10 carbon atoms, and preferably of 1 to about 8 carbon atoms, and X represents either H or R as described above. The viscosity at 25°C of the organohydrogenpolysiloxane of the constituent (C) is within a range from 0.001 to 1.0 Pa·s, with values from 0.01 to 0.1 Pa·s being preferred.

[0015]

The total number of hydrogen atoms bonded to silicon atoms (namely, SiH groups) within the aforementioned constituent (B) and constituent (C) is in a range of 1 to 5 atoms, and preferably 1 to 3 atoms, per alkenyl group within the aforementioned constituent (A). Furthermore, the number of hydrogen atoms bonded to silicon atoms within the constituent (B) accounts for 20 to 70 mol%, and preferably 30 to 60 mol%, of the total number of hydrogen atoms bonded to silicon atoms within the constituent (B) and the constituent (C).

[0016]

[Constituent (D)]

In the present invention, any catalyst which promotes the addition reaction (hydrosilylation) between the alkenyl groups of the constituent (A) and the SiH groups within the constituents (B) and (C) may be used as the hydrosilylation reaction catalyst of the constituent (D), and any of the

catalysts commonly used for such purposes can be used. For example, at least one catalyst selected from the group consisting of platinum based catalysts, palladium-based catalyst based catalysts and rhodium based catalysts may be used, and specific examples include chloroplatinic acid, alcohol modified products of chloroplatinic acid, coordination compounds of chloroplatinic acid with olefins, vinyl siloxanes or acetylene compounds, tetrakis(triphenylphosphine)palladium and chlorotris(triphenylphosphine)rhodium, although platinum based compounds are particularly desirable. The constituent (D) should be used in an effective quantity as a catalyst (so-called catalytic quantity) and specifically incorporated so that for example, the quantity of the catalyst (in terms of the quantity of the metallic element) should be 0.01 to 500 ppm, and preferably 0.1 to 100 ppm by weight, relative to the combined weight of the constituents (A), (B) and (C).

[0017]

[Constituent (E)]

In the present invention, the finely powdered silica of the constituent (E) functions as a reinforcing agent. A composition of the present invention is particularly suited for use as a mold material, and the cured product is molded into various shapes as a matrix. Consequently, the cured product may be formed into a reverse taper shaped matrix, in which case the strength characteristics of the matrix, and particularly the tear strength, are important, and by using finely powdered silica as a reinforcing agent in compositions of the present invention, it becomes possible to form cured products capable of satisfying these types of strength requirements. The finely powdered silica must

have a specific surface area of at least 50 m<sup>2</sup>/g, as measured by BET methods, with values of 100 to 300 m<sup>2</sup>/g being preferred. At specific surface area values less than 50 m<sup>2</sup>/g, satisfactory strength characteristics cannot be obtained.

[0018]

Examples of this type of finely powdered silica, which display a specific surface area within the aforementioned range, include synthetic silica, e.g., dry process silica such as fumed silica and wet process silica. These types of silica have a large number of silanol groups on the surface, and so may also be used as so-called treated silica, where the surface is treated with a halogenated silane, an alkoxy silane, various silazane compounds, or the like. Furthermore, the amount of this finely powdered silica incorporated should be no more than 50 parts by weight per 100 parts by weight of the organopolysiloxane of the constituent (A), with quantities of 10 to 40 parts by weight being preferred, and it is properly incorporated within this range so as to achieve an appropriate degree of strength. If the amount incorporated exceeds 50 parts by weight per 100 parts by weight of the constituent (A), then the workability of the composition deteriorates.

[0019]

[Constituent (F)]

In the present invention the constituent (F) is optionally used where necessary as an internal mold releasing agent for the composition, and should preferably be a straight chain non-functional organopolysiloxane. If this organopolysiloxane were to have functional groups capable of undergoing addition reactions such as alkenyl groups or hydrosilyl groups (SiH groups), then it would

become immobilized within the cured rubber, and provide no effect in reducing the release force. Consequently, examples of suitable substituent groups for bonding to the silicon atoms of this non-functional organopolysiloxane include alkyl groups such as methyl groups, ethyl groups and propyl groups, aryl groups such as phenyl groups and tolyl groups, and halosubstituted alkyl groups such as 3,3,3-trifluoropropyl groups and 3-chloropropyl groups. The viscosity at 25°C of the non-functional organopolysiloxane is within a range from 0.01 to 500 Pa·s, with values of 0.03 to 100 Pa·s being preferred. The amount of this constituent incorporated should be from 0 to 20 parts by weight, and preferably 5 to 10 parts by weight, per 100 parts by weight of the constituent (A). If the amount of the constituent (F) incorporated exceeds 20 parts per 100 parts by weight of the constituent (A), then oil bleeding increases, and the transference of silicone to the resin replica products becomes increasingly likely.

[0020]

[Other constituents]

In a composition of the present invention, in addition to the constituents described above, other known extenders may also be added, provided the effects of the present invention in improving the mold release durability are not impaired. Examples of such extenders include organopolysiloxane resins incorporating  $\text{SiO}_2$  units or  $\text{R}^1\text{SiO}_{3/2}$  units and with at least two alkenyl groups in a single molecule. Furthermore, reaction controlling agents may also be used, and any of the conventional addition reaction controlling agents may be used, including acetylene based compounds such as 3-methyl-1-butyne-3-ol, 3,5-dimethyl-1-hexyne-3-ol and phenylbutynol;

alkenylsiloxanes such as 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane and 1,3-divinyl-tetramethyldisiloxane; triazole compounds such as benzotriazole; as well as phosphine compounds and mercapto compounds, any of which can be added in minute or small quantities. In addition, inorganic pigments such as cobalt blue; coloring agents such as organic dyes; and additives for improving the heat resistance and fire resistance such as cerium oxide, zinc carbonate, manganese carbonate, red iron oxide, titanium oxide and carbon black may also be added.

[0021]

[Examples]

As follows is a specific description of the present invention using examples, although the present invention is in no way limited to these examples.

[0022]

#### Example 1

A mixture of: (a) 35 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyltrimethylsilyl group and having a viscosity at 25°C of approximately 1 Pa·s, (a') 30 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyltrimethylsilyl group and having a viscosity at 25°C of approximately 0.4 Pa·s, 20 parts by weight of hydrophobic silica with a specific surface area of 120 m<sup>2</sup>/g which had been treated with trimethylsilyl groups, and 25 parts by weight of powdered quartz with an average particle diameter of 5 μm was placed in a kneader, and with the mixture well stirred, 5 parts by weight of hexamethylenedisilazane and 2.5 parts by weight of water were added, and the resulting mixture was

then stirred for 1 hour without heating. The temperature was then raised to 150°C, and the mixing continued for a further 2 hours, before the temperature was cooled to room temperature. To 100 parts by weight of the thus obtained mixture were added: (b) 4.0 parts by weight of a dimethylpolysiloxane with a hydrogen atom bonded to a silicon atom (SiH group) at both terminals of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.13 weight %) and having a viscosity at 25°C of 0.018 Pa·s, (c) 2.3 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.38 weight %) and having a viscosity at 25°C of 0.015 Pa·s, 4 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked with a trimethylsilyl group and having a viscosity at 25°C of approximately 100 Pa·s, sufficient quantity of a complex of chloroplatinic acid and divinyltetramethyldisiloxane to provide 30 ppm of platinum metal relative to the combined quantity of the aforementioned constituents (a), (a'), (b) and (c), and 0.1 parts by weight of 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane as a controlling agent, and following careful stirring of the mixture, the mixture was degassed under vacuum. The mixture was then cured for 2 hours at 60°C, and used to prepare a sheet in accordance with JIS K 6249, and the general properties of the sheet were then measured.

[0023]

A concave shaped matrix was also formed in the same manner. A urethane resin (3017 manufactured by H & K Corporation) was then poured into this concave shaped mold,



and molding was carried out by curing the urethane resin over a one hour period at 70°C. This molding operation was repeated and, until the urethane resin adhered to the silicone mold, the mold durability was evaluated. The general properties, and the results of the mold durability evaluation are shown in Table 1.

[0024]

Example 2

A mixture of: (a) 65 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyl dimethylsilyl group and having a viscosity at 25°C of approximately 1 Pa·s, 20 parts by weight of hydrophobic silica with a specific surface area of 120 m<sup>2</sup>/g which had been treated with trimethylsilyl groups, and 35 parts by weight of powdered quartz with an average particle diameter of 5 μm was placed in a kneader, and with the mixture well stirred, 5 parts by weight of hexamethylenedisilazane and 2.5 parts by weight of water were added, and the resulting mixture was then stirred for 1 hour without heating. The temperature was then raised to 150°C, and the mixing continued for a further 2 hours, before the temperature was cooled to room temperature. To 100 parts by weight of the thus obtained mixture were added: (b) 3.5 parts by weight of a dimethylpolysiloxane with a hydrogen atom bonded to a silicon atom at both terminals of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.13 weight %) and having a viscosity at 25°C of 0.018 Pa·s, (c) 1.4 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen

atoms = 0.38 weight %) and having a viscosity at 25°C of 0.15 Pa·s, 5 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked with a trimethylsilyl group and having a viscosity at 25°C of approximately 100 Pa·s, sufficient quantity of a complex of chloroplatinic acid and divinyltetramethyldisiloxane to provide 30 ppm of platinum metal relative to the combined quantity of the aforementioned constituents (a), (b) and (c), and 0.1 parts by weight of 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane as a controlling agent, and following careful stirring of the mixture, the mixture was degassed under vacuum. The mixture was then cured for 2 hours at 60°C, and used to prepare a sheet in accordance with JIS K 6249, and the general properties of the sheet were then measured. The mold durability was also evaluated in the same manner as the Example 1, and these results are shown in Table 1.

[0025]

#### Example 3

A mixture of: (a) 65 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyltrimethylsilyl group and having a viscosity at 25°C of approximately 1 Pa·s, 20 parts by weight of hydrophobic silica with a specific surface area of 120 m<sup>2</sup>/g which had been treated with trimethylsilyl groups, and 35 parts by weight of powdered quartz with an average particle diameter of 5 μm was placed in a kneader, and with the mixture well stirred, 5 parts by weight of hexamethylenedisilazane and 2.5 parts by weight of water were added, and the resulting mixture was then stirred for 1 hour without heating. The temperature was then raised to

150°C, and the mixing continued for a further 2 hours, before the temperature was cooled to room temperature. To 100 parts by weight of the thus obtained mixture were added: (b) 6.9 parts by weight of a dimethylpolysiloxane with a hydrogen atom bonded to a silicon atom at both terminals of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.07 weight %) and having a viscosity at 25°C of 0.04 Pa·s, (c) 1.3 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.38 weight %) and having a viscosity at 25°C of 0.15 Pa·s, sufficient quantity of a complex of chloroplatinic acid and divinyltetramethyldisiloxane to provide 30 ppm of platinum metal relative to the combined quantity of the aforementioned constituents (a), (b) and (c), and 0.1 parts by weight of 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane as a controlling agent, and following careful stirring of the mixture, the mixture was degassed under vacuum. The mixture was then cured for 2 hours at 60°C, and used to prepare a sheet in accordance with JIS K 6249, and the general properties of the sheet were then measured.

Furthermore, the mold durability was also evaluated in the same manner as the Example 1, and these results are shown in Table 1.

[0026]

#### Example 4

A mixture of: (a) 65 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyltrimethylsilyl group and having a viscosity at 25°C of approximately 1 Pa·s, 20 parts by

weight of dried silica with a specific surface area of 200 m<sup>2</sup>/g. and 35 parts by weight of powdered quartz with an average particle diameter of 5  $\mu$ m was placed in a kneader, and with the mixture well stirred, 5 parts by weight of hexamethylenedisilazane and 2.5 parts by weight of water were added, and the resulting mixture was then stirred for 1 hour without heating. The temperature was then raised to 150°C, and the mixing continued for a further 2 hours, before the temperature was cooled to room temperature. To 100 parts by weight of the thus obtained mixture were added: (b) 3.5 parts by weight of a dimethylpolysiloxane with a hydrogen atom bonded to a silicon atom at both terminals of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.13 weight %) and having a viscosity at 25°C of 0.18 Pa·s, (c) 1.4 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms at a terminal of the molecular chain and on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.38 weight %) and having a viscosity at 25°C of 0.15 Pa·s, 5 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked with a trimethylsilyl group and having a viscosity at 25°C of approximately 100 Pa·s, sufficient quantity of a complex of chloroplatinic acid and divinyltetramethyldisiloxane to provide 30 ppm of platinum metal relative to the combined quantity of the aforementioned constituents (a), (b) and (c), and 0.1 parts by weight of 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane as a controlling agent, and following careful stirring of the mixture, the mixture was degassed under vacuum. The mixture was then cured for

2 hours at 60°C, and used to prepare a sheet in accordance with JIS K 6249, and the general properties of the sheet were then measured. The mold durability was also evaluated in the same manner as the Example 1, and these results are shown in Table 1.

[0027]

Example 5

A mixture of: (a) 100 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyl dimethylsilyl group and having a viscosity at 25°C of approximately 1 Pa·s, 15 parts by weight of hydrophobic silica with a specific surface area of 120 m<sup>2</sup>/g which had been treated with trimethylsilyl groups, and 30 parts by weight of wet silica with a specific surface area of 200 m<sup>2</sup>/g was placed in a kneader, and with the mixture well stirred, 5 parts by weight of hexamethylenedisilazane and 2.5 parts by weight of water were added, and the resulting mixture was then stirred for 1 hour without heating. The temperature was then raised to 150°C, and the mixing continued for a further 2 hours, before the temperature was cooled to room temperature. To 100 parts by weight of the thus obtained mixture were added: (b) 4.5 parts by weight of a dimethylpolysiloxane with a hydrogen atom bonded to a silicon atom at both terminals of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.13 weight %) and having a viscosity at 25°C of 0.018 Pa·s, (c) 1.5 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.4 weight %) and having a viscosity at 25°C of 0.01 Pa·s, sufficient quantity of a complex of

chloroplatinic acid and divinyltetramethyldisiloxane to provide 30 ppm of platinum metal relative to the combined quantity of the aforementioned constituents (a), (b) and (c), and 0.1 parts by weight of 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane as a controlling agent, and following careful stirring of the mixture, the mixture was degassed under vacuum. The mixture was then cured for 2 hours at 60°C, and used to prepare a sheet in accordance with JIS K 6249, and the general properties of the sheet were then measured. The mold durability was also evaluated in the same manner as the Example 1, and these results are shown in Table 1.

[0028]

#### Example 6

A mixture of: (a) 93 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyltrimethylsilyl group and having a viscosity at 25°C of approximately 5 Pa·s, (a') 7 parts by weight of an organopolysiloxane resin comprising 39.5 mol% of  $(\text{CH}_3)_3\text{SiO}_{1/2}$  units, 6.5 mol% of  $(\text{CH}_3)_2(\text{CH}_2=\text{CH})\text{SiO}_{1/2}$  units, and 54 mol% of  $\text{SiO}_2$  units, and 25 parts by weight of hydrophobic silica with a specific surface area of 120 m<sup>2</sup>/g which had been treated with trimethylsilyl groups was placed in a kneader, and with the mixture well stirred, 5 parts by weight of hexamethylenedisilazane and 2.5 parts by weight of water were added, and the resulting mixture was then stirred for 1 hour without heating. The temperature was then raised to 150°C, and the mixing continued for a further 2 hours, before the temperature was cooled to room temperature. To 100 parts by weight of the thus obtained mixture were added: (b) 6 parts by weight of a dimethylpolysiloxane with a hydrogen atom bonded to a

silicon atom at both terminals of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.13 weight %) and having a viscosity at 25°C of 0.018 Pa·s, (c) 0.7 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms at a terminal of the molecular chain and on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.48 weight %) and having a viscosity at 25°C of 0.012 Pa·s, sufficient quantity of a complex of chloroplatinic acid and divinyltetramethyldisiloxane to provide 30 ppm of platinum metal relative to the combined quantity of the aforementioned constituents (a), (a'), (b) and (c), and 0.1 parts by weight of 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane as a controlling agent, and following careful stirring of the mixture, the mixture was degassed under vacuum. The mixture was then cured for 2 hours at 60°C, and used to prepare a sheet in accordance with JIS K 6249, and the general properties of the sheet were then measured. Furthermore, the mold durability was also evaluated in the same manner as the Example 1, and these results are shown in Table 1.

[0029]

#### Comparative Example 1

A mixture of: (a) 65 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked by a vinyl dimethylsilyl group and having a viscosity at 25°C of approximately 1 Pa·s, 20 parts by weight of hydrophobic silica with a specific surface area of 120 m<sup>2</sup>/g which had been treated with trimethylsilyl groups, and 15 parts by weight of powdered quartz with an average particle diameter of 5 μm was placed in a kneader,

and with the mixture well stirred, 5 parts by weight of hexamethylenedisilazane and 2.5 parts by weight of water were added, and the resulting mixture was then stirred for 1 hour without heating. The temperature was then raised to 150°C, and the mixing continued for a further 2 hours, before the temperature was cooled to room temperature. To 100 parts by weight of the thus obtained mixture were added (b) 0.67 parts by weight of a dimethylpolysiloxane with a hydrogen atom bonded to a silicon atom at both terminals of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.26 weight %) and having a viscosity at 25°C of 0.005 Pa·s, (c) 1.84 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms at a terminal of the molecular chain and on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.54 weight %) and having a viscosity at 25°C of 0.012 Pa·s, 5 parts by weight of a dimethylpolysiloxane with both terminals of the molecular chain blocked with a trimethylsilyl group and having a viscosity at 25°C of approximately 100 Pa·s, sufficient quantity of a complex of chloroplatinic acid and divinyltetramethyldisiloxane to provide 30 ppm of platinum metal relative to the combined quantity of the aforementioned constituents (a), (b) and (c), and 0.1 parts by weight of 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane as a controlling agent, and following careful stirring of the mixture, the mixture was degassed under vacuum. The mixture was then cured for 2 hours at 60°C, and used to prepare a sheet in accordance with JIS K 6249, and the general properties of the sheet were then measured. Furthermore, the mold durability was



also evaluated in the same manner as the Example 1, and these results are shown in Table 2.

[0030]

#### Comparative Example 2

Using 100 parts by weight of a compound mixture prepared in the Example 1 without the powdered quartz with an average particle diameter of 5  $\mu\text{m}$ , a composition was prepared according to the Example 1, with the exceptions of not using the constituent (B), but using 3.1 parts by weight of a methylhydrogenpolysiloxane with hydrogen atoms bonded to silicon atoms at a terminal of the molecular chain and on side chains of the molecular chain (wherein the content of silicon atom bonded hydrogen atoms = 0.54 weight %) and having a viscosity at 25°C of 0.012 Pa's (12  $\text{mm}^2/\text{s}$ ), and the thus prepared composition was cured and used to prepare a sheet, and the general properties of the sheet were then measured. Furthermore, the mold durability was also evaluated in the same manner as the Example 1, and these results are shown in Table 2.

[0031]

[Table 1]

|  | Ex. 1 | Ex. 2 | Ex. 3 | Ex. 4 | Ex. 5 | Ex. 6 |
|--|-------|-------|-------|-------|-------|-------|
| Hardness<br>(type A)                           | 35    | 38    | 40    | 38    | 25    | 20    |
| Elongation<br>at shearing<br>(%)               | 480   | 470   | 450   | 460   | 600   | 850   |
| Tensile<br>strength<br>(MPa·s)                 | 5.4   | 5.3   | 4.9   | 5.3   | 7.0   | 5.0   |
| Tear<br>strength<br>(kN/m)                     | 17    | 22    | 22    | 21    | 25    | 10    |
| Rate of<br>linear<br>cross-<br>linking<br>(%)* | 37    | 46    | 49    | 46    | 49    | 70    |
| Mold<br>release<br>repetitions<br>(times)      | 65    | 70    | 70    | 70    | 75    | 65    |

\* The ratio (mol%) of SiH groups in the constituent (B) relative to the total of SiH groups in the combined constituent (B) and constituent (C)

[0032]

[Table 2]

|  | Comp. EX 1 | Comp. EX. 2 |
|--|------------|-------------|
| Hardness<br>(type A)                     | 40         | 40          |
| Elongation at<br>shearing (%)            | 410        | 410         |
| Tensile<br>strength<br>(MPa's)           | 5.0        | 5.0         |
| Tear strength<br>(kN/m)                  | 12         | 8           |
| Rate of<br>linear cross-<br>linking (%)* | 15         | -           |
| Mold release<br>repetitions<br>(times)   | 45         | 30          |

\* The ratio (mol%) of SiH groups in the constituent (B) relative to the total of SiH groups in the combined constituent (B) and constituent (C)

[0033]

[Effects of the Invention]

An organopolysiloxane composition for molding purposes according to the present invention displays superior mold releasability relative to urethane resins, epoxy resins, dicyclopentadiene resins, polyester resins, and the like, and moreover also displays superior elongation at shearing and tear strength and can be suitably used as a highly durable mold composition.